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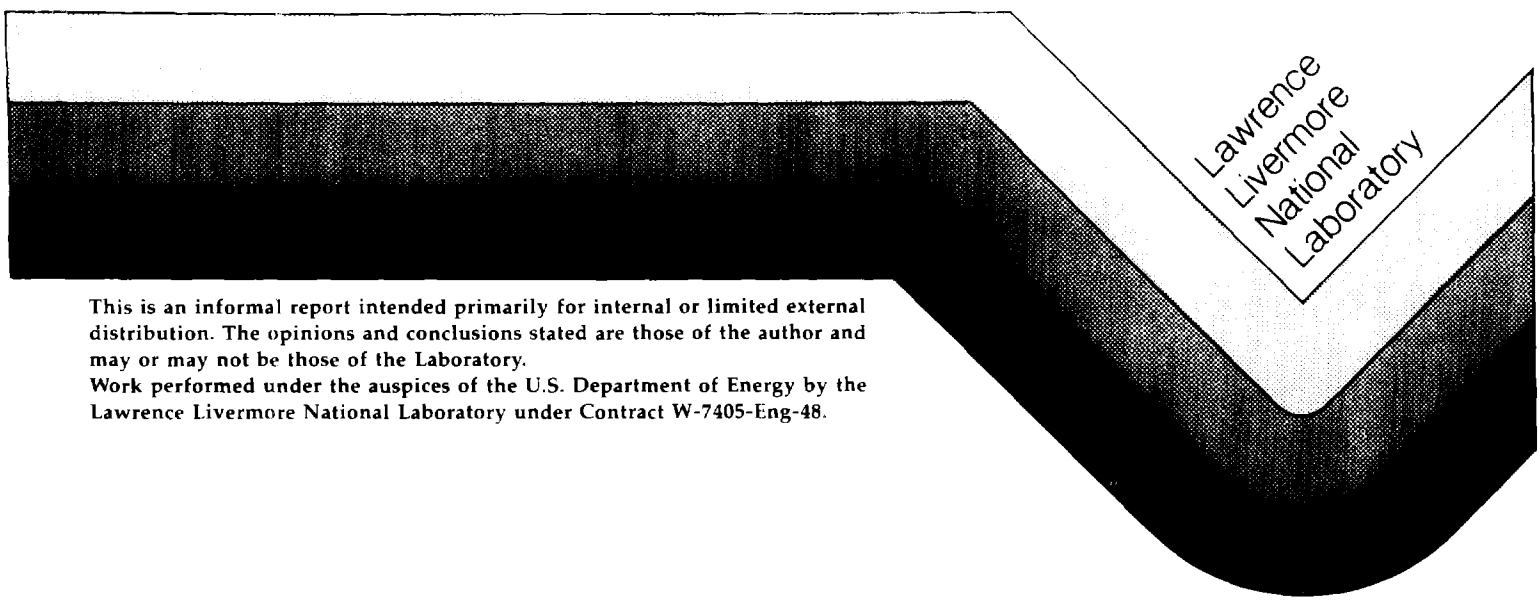
FUSION MATERIALS SCIENCE AT REACTOR 14-MeV NEUTRON FLUXES: UPGRADING RTNS TARGETS INTO THE MULTI-MEGAWATT/M^2 REGIME

David B. Tuckerman

15 March 1984



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1 Summary

The Rotating Target Neutron Source-II (RTNS-II) facility is the most intense continuous source of 14-MeV neutrons in the world. It is used to study the effects of fast neutrons on materials, to determine their suitability for use in fusion reactors. In RTNS-II, a water-cooled rotating target coated with titanium tritide is bombarded with deuterons. A small fraction of the incident deuterons fuse with the tritons in the target, producing 14.3-MeV neutrons. At present the neutron flux is substantially less than what a fusion test reactor would generate. This report examines the possibilities for upgrading RTNS targets to produce reactor-level neutron fluxes (or more). It is shown that the existing targets are operating near their thermal limit. However, modifications in target design and operating conditions are possible which could reasonably support up to a 30-fold increase in peak neutron flux ($\sim 3 \times 10^{14}$ neutrons/cm^2-sec, or 6 MW/m^2). The irradiated volume could also be increased, if desired. It seems likely that with some research and experimentation with palladium underlayers, target cladding/overcoating, and/or in-situ retritiding, an acceptable target lifetime can still be achieved at this greatly upgraded neutron flux. The proposed target modifications consist of a number of significant incremental improvements on the existing system, rather than one large breakthrough. Some of them could be implemented rapidly (time scale of less than a year); others would require somewhat more research (time scale of 2 or 3 years, depending on funding and staffing levels, and difficulties encountered). Each change can be independently omitted should technological difficulties arise. As such the overall RTNS upgrade process would be low-risk and high-payoff.

2 Present RTNS Target Performance

The present RTNS-II target system is shown in Figure 1. The target is an internally cooled copper disk of radius 25 cm, formed into a spherical segment and bonded to a conical vacuum chamber. A rotating vacuum seal [1] is located at the apex of the cone so that the entire target chamber can rotate at 5000 RPM. A bellows enables the target to be slowly tilted so that the entire usable target area (from a radius of R=10 cm out to R=25 cm) is eventually scanned by the deuteron beam. Chilled water is fed to the target through a rotating seal at the center of the target. The target cross section (Figure 2) consists of a water-cooled copper alloy, electroplated with 500 μm of copper, and coated with 10 μm of titanium tritide (TiT_x, where initially $x \simeq 1.8$) [2]. The target is bombarded with deuterons of 375 keV energy at currents of up to 125 mA; hence 47 kW of heat is dissipated in the target. The beam shape is approximately Gaussian ($\exp -r^2/2r_o^2$). If its FWHM is 1 cm ($r_o = .425$ cm), this gives a peak heat flux of 41 kW/cm^2 at the beam center. Actually the larger (50-cm) targets are suspected [3] to give wider beams (FWHM of

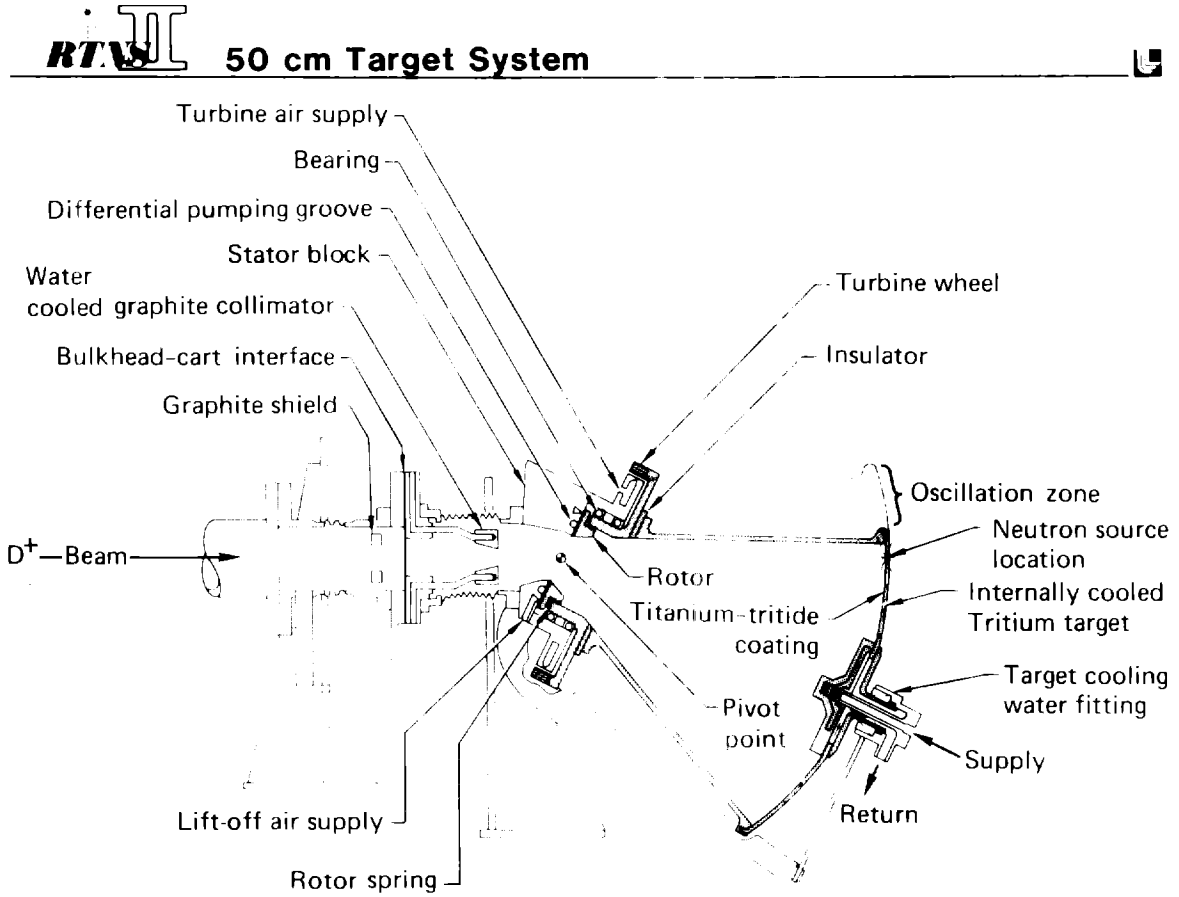

Figure 1: The RTNS target system (figure courtesy of C. M. Logan).

~1.5 cm), but I shall assume for purposes of this analysis that this could be tightened to 1 cm by fine-tuning the ion optics. Up to 4.2×10^{-5} neutrons are produced per incident deuteron in a fresh target [4]. Thus, at the surface of the tritide, a fresh target would yield a 14.3-MeV neutron output of 3.28×10^{13} neutrons/sec, for a neutron flux of 1.44×10^{13}/cm^2-sec in each direction from the beam center, or 0.34 MW/m^2. This is not far below fusion reactor first-wall neutron fluxes, which are expected to be in the 1-10 MW/m^2 regime. However due to the intervening copper, target runout, and running clearance, samples must be placed at least 0.35 cm away from the tritide neutron source [5]. Since this is comparable to the spot radius, a significant reduction in flux results, to about 0.9×10^{13}/cm^2-sec over the beam center (or 0.21 MW/m^2).

The peak temperature of the RTNS rotating target has never been directly measured. However, if we assume that no barrier to tritium outgassing exists, we may infer from the fact that the targets last ~100 hours that it does not exceed 300°C at any time. (There is some evidence [6] that this is not an entirely correct assumption; a thin oxide layer may indeed reduce the tritium outgassing rate. This would increase the allowable peak temperature T_{peak} somewhat, but probably not as much as could be achieved by

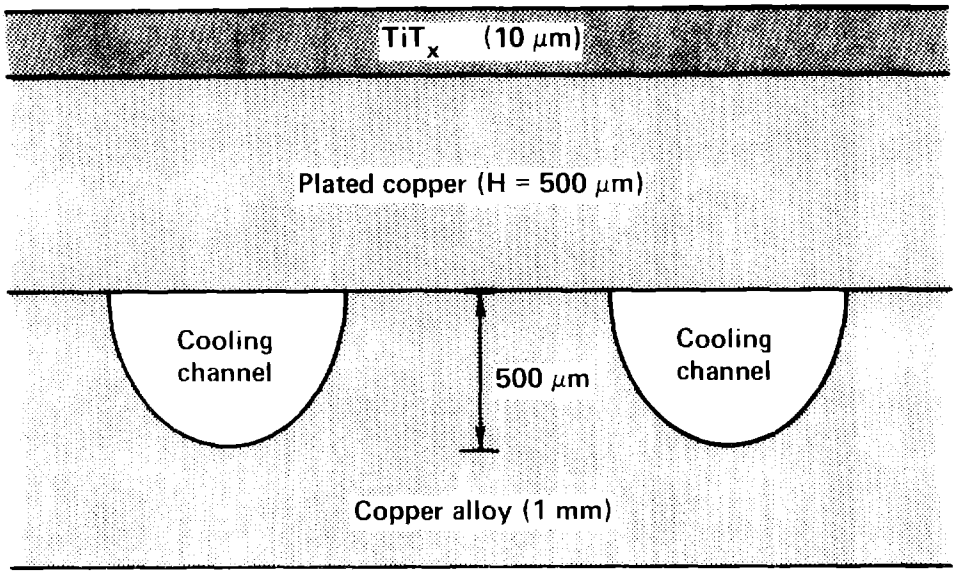

Figure 2: A typical cross section of the present RTNS-II targets.

cladding the TiT_x with a diffusion barrier, as discussed later.) This is because the dissociation pressure of TiH_x is $P_d \simeq (1.45\times10^{12})\cdot e^{-19600/T}$ Torr for $1\leq x\leq 1.5$; for $x>1.5$ the dissociation is much more rapid (Figure 3). The tritium flux would be approximately $P_d(2\pi mkT)^{-1/2} = A\cdot e^{-19600/T}$, where $A=3.5\times10^9$ mol/cm^2-sec. By expanding the target surface temperature T(R) about R=0 this can be integrated over the spot area to yield a total hydrogen loss rate of

$$2\pi r_o^2 A[T_{peak}^2/19600(T_{peak}-T_A)]\ \exp(-19600/T_{peak})\ \text{mol/sec.}$$

(T_A is the ambient temperature, which we take to be 300°K.) For the present RTNS target's TiT_x thickness of 10 μm (4.2 mg/cm^2) and its beam-scanned area of $\pi(25^2-10^2)=1649$ cm^2, the tritium content would decay exponentially with a time constant of 106 hours if $T_{peak}=300$°C. There is convincing evidence [6, 8] that under the present operating conditions the target lifetime is limited by deuterium displacement of the tritium, but this argument is still valid for establishing an **upper bound** on the peak temperature T_{peak} of the present RTNS target.

2.1 Temperature Rise in the TiT_x Layer

The electrical and thermal properties of titanium tritide are not expected to differ significantly from that of titanium hydride. Unfortunately, there appears to be no data in the literature on the thermal conductivity of either TiT_x or TiH_x in the form used on RTNS targets, nor have any such data been taken at LLNL. Data exist for powdered $TiMn_{.5}$ and FeTi hydrides [9, 10] showing $k\simeq0.01$-0.015 W/cm-K, but these data are useless because here the thermal conductivity is dominated by thermal contact resistance between the particles. Data on thermal conductivity do exist for massive (i.e. not cracked or powdered) zirconium hydride. The bulk thermal conductivity of ZrH_x was measured to be 0.20 W/cm-K

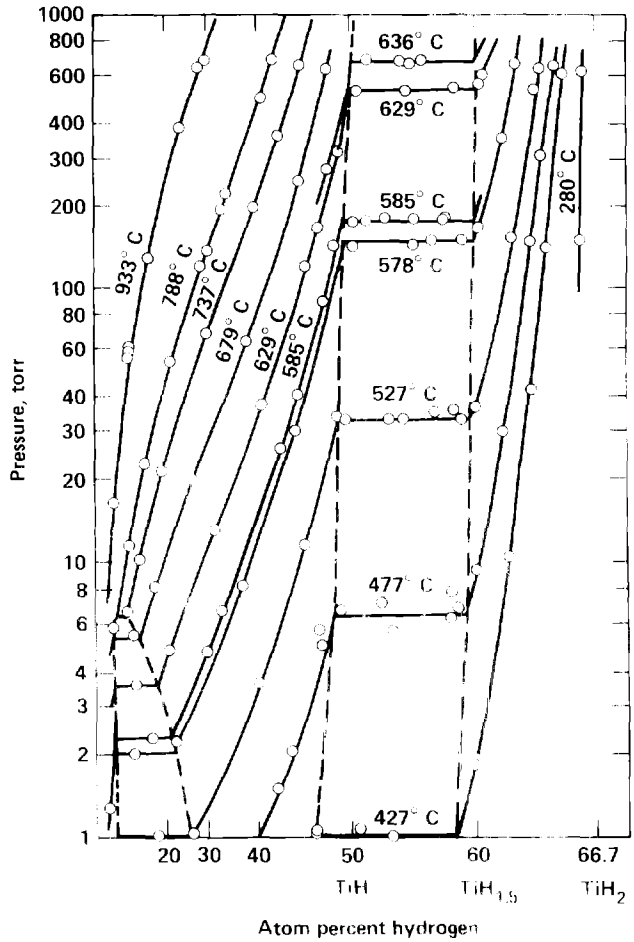

Figure 3: Pressure-temperature-composition relations for the titanium-hydrogen system (from Ref. [7])

at 400°K; this is the same as that of bulk Zr metal! The thermal conductivity of titanium metal is also 0.20 W/cm-K, so by appeal to its congener relation to Zr (both Zr and Ti are Group 4B transition elements), I expect TiT_x to have a similar bulk thermal conductivity. This is not a very high value for a metal (compare with copper, k=4 W/cm-K). In RTNS targets, a 10-μm layer of Ti is electron-beam evaporated onto a copper alloy cold plate. The targets are then tritided by exposure to tritium gas at 350°C. According to Steve Steward of LLNL, who has performed this process, the surface does not appear cracked or powdered in any way, but retains its metallic luster and shiny appearance. Thus we expect to achieve the bulk thermal conductivity in this TiT_x layer.

The specific heat of TiH_x (presumably the granularity is irrelevant here) has been measured to be C≃0.8 J/gm-K at 400°K [11].

The path length and mean projected (forward) range of a deuteron in titanium **metal** is given in

Ref. [12]. At 375 keV, the range is 2.4 μm. The stopping is primarily due to many coulomb interactions with electrons (coulomb interactions with nuclei are negligible by comparison); thus the standard deviation is small (about 10% at the energies under consideration). So all deuterons have more or less the same energy loss profile. The effect of the bound tritium in TiT_x could probably be well approximated by simply adding the stopping power of its tritium fraction to that of the titanium (Bragg's Law [13]). This is because the additional electron will either remain bound as a low-energy valence electron or it will be donated to the conduction d-band of the metal; in either case the electron will not be as highly localized as those in the titanium core levels. Thus it will be relatively effective in its stopping power. Assuming a 10% volume expansion in the Ti thin film due to the tritiding process, my estimate of the 375-keV deuteron projected range in $TiT_{1.5}$ is 1.87 μm, as tabulated in Table 1 (compare with 2.4 μm for pure Ti metal).

The energy loss $\delta E/\delta x$ for electronic stopping is roughly constant at these energies, thus the generated heat flux is approximately uniform throughout the 1.87 μm stopping region. The thermal diffusivity of TiT_x is $\alpha=k/\rho C=0.0625$ cm^2/sec. The exponential penetration depth z_o of heat into a material of thermal diffusivity α generated by a moving Gaussian beam $\exp(-r^2/2r_o^2)$ is known [14] to be $z_o \simeq 1.7(\alpha r_o/V)^{1/2}$, where $r_o=0.425$ cm for a FWHM of 1.0 cm. At a point 80% of the way out to the spinning target's circumference, $V \simeq 1.0\times10^4$ cm/sec, and thus $z_o=28$ μm for TiT_x. This is substantially greater than the actual TiT_x thickness (10 μm); hence this is nearly a steady-state heat flux problem as far as the TiT_x layer is concerned, which is unfortunate as it implies that the target rotation is not helping at all in thermal management in this layer. The thermal resistance R (cm^2-°C/W) of this layer is thus $R=[R_p/2 + (t-R_p)]/k_{TiT_x}$; the factor of 1/2 comes from the fact that the heat generation is distributed uniformly over a depth R_p, hence the heat source has its centroid at a depth $R_p/2$, rather than being localized at the surface. For a deuteron range of $R_p=1.87$ μm and a TiT_x thickness of t=10 μm we have R=4.5 m^2-°C/kW. The present peak flux of 11 kW/cm^2 thus implies $\Delta T_{tritide}=185$°C in the TiT_x layer alone!

2.2 Temperature Rise In the Copper Substrate

The peak temperature rise as a function of depth induced by a scanning Gaussian beam of total power q and velocity V in a bulk material of thermal conductivity k and volumetric heat capacity ρC can be approximated [14] by $\Delta T_{Cu}(z)=0.235q(k\rho C r_o^3 V)^{1/2}\exp(-z/z_o)$, where z_o is the penetration depth defined previously. The thermal conductivity of copper is k=4.0 W/cm-K; its heat capacity is $\rho C=3.83$ J/cm^3-K. For the 47 kW, 1 cm^2 FWHM beam, we have $\Delta T_{Cu}=89$°C at the rim of the target (R=25 cm). However, for target longevity it is necessary to use more area than the single beam-width band right at the rim; presumably one would scan the deuteron beam over at least the band from R=20 cm to R=25 cm. At R=20 cm, we have $V=1.05\times10^4$ cm/sec, hence $\Delta T_{Cu}=99$°C.

The preceding analysis neglected the heat capacity of the TiT_x layer. The TiT_x is at an average temperature of $\Delta T_{Cu}+\Delta T_{tritide}/2$, hence the heat stored in this layer is $Cm''(\Delta T_{Cu}+\Delta T_{tritide}/2)$ where C and m'' are the heat capacity (J/gm-K) and mass (gm/cm^2) of the tritide layer. Subtracting this from the peak beam energy of 4.2 J/cm^2 leads to $\Delta T_{Cu}=84°C$.

2.3 Convective Heat Transfer

The RTNS targets contain cooling channels of depth 0.5 mm and widths of 0.5 to 1.0 mm. Thus their aspect ratio is less than unity, so the heat-transfer surface area is approximately equal to the target surface area. The channels meander somewhat in an attempt to increase the turbulent heat transfer, but this approach is based on achieving fairly ideal "surface spoilers" [15], which is probably not the case for the chemically etched channels; the actual convective heat transfer in RTNS targets has never been measured. In the worst case of effectively smooth channels, the present design would have a Reynolds number of approximately 6500, a Colburn number of $j_H=.004$, and hence a Nusselt number of $Nu=j_H RePr^{1/3}=50$. In the best case of optimally roughened channels, we would have Re=4500, $j_H=.009$, and hence $Nu\simeq 80$. For the channel hydraulic diameter of .05 cm, these correspond to heat transfer coefficients of h=6.0 to 9.4 W/cm^2-K. I will take an intermediate value and assume that h=7.5 W/cm^2-K.

The plated copper acts as a thermal capacitor: it is charged by a rapid heat pulse as the beam sweeps over it, then it slowly discharges its heat into the cooling channels. Immediately after the pulse, the temperature distribution in the copper decays exponentially with depth, where the penetration depth is $z_o=1.7(\alpha r_o/V)^{1/2}$ [14]. In the case of the plated copper, we have $\alpha=(4.0\ W/cm\text{-}K)/(3.83\ J/cm^3\text{-}K)=1.04\ cm^2/s$, so $z_o=110\ \mu m$ at R=20 cm. After the beam has passed, the heat will redistribute itself uniformly within the copper bulk; this occurs with a time constant $\tau_{Cu}=(H^2/\alpha)$, where H is the thickness of the copper between the tritide and the channels. At present, H is 500 μm; however, an additional 1 mm of copper thickness exists in the region between the channels, so the effective thickness of the copper is more like 1 mm. Thus $\tau_{Cu}=9.6$ msec. The target rotation period is 12 msec, so this thermal redistribution process is not complete by the time the beam again sweeps over the reference spot.

The discharge of the heat into the cooling channels is an even slower process; its thermal time constant is

$$\tau_{convective}=(\rho CH)/h=(3.83\ J/cm^3\text{-}K)(0.1\ cm)/(7.5\ W/cm^2\text{-}K)=51\ msec.$$

which is much greater than the rotation period. So the copper underlying the TiT_x layer discharges very little of its stored heat over a full rotation of the target. The copper will therefore attain a nearly constant bulk temperature rise $\Delta T_{conv}=\langle q''\rangle/h$ throughout its thickness, where $\langle q''\rangle$ is the time-averaged heat flux and h is the convective heat-transfer coefficient. $\langle q''\rangle$ is simply the peak flux

divided by $(2\pi)^{1/2}(R/r_o)$; at R=20 cm this is a factor of 118. Thus ΔT_{conv}=(350 W/cm^2)/(7.5 W/cm^2-K)=47°C. In addition, there will be a caloric temperature rise due to heating of the water. At a flow rate of 34 l/m and a 47 kW beam, the output water will be heated 20°C. The channels follow a "hairpin" path, going from R=10 cm out to R=25 cm and back again. We are only concerned with the caloric temperature rise at the peak temperature of the substrate, which occurs at the center of the beam on the **downstream** channels. At this point 3/4 of the heat has been deposited into the coolant, so $\Delta T_{caloric}$=15°C.

2.4 Summary of Present Target Thermal Performance

At R=20 cm (80% of the distance from the rotating target's center to its rim), the following temperature rises are calculated

TiT_x (10 μm thick):	$\Delta T_{tritide}$ =	185°C
Copper alloy (k≃3.4 W/cm-K):	ΔT_{Cu}=	84°C
Water (boundary layer)	ΔT_{conv}=	47°C
Water (caloric heating)	$\Delta T_{caloric}$=	15°C
Total:	ΔT=	331°C

Assuming an input chilled water temperature of 4°C, the peak surface of the target is presently in the neighborhood of T_{peak}=335°C.

This presents a puzzle, in that the previous calculations indicate that the targets would not last 100 hours if T_{peak}>300°C. There are three possible solutions to this.

First, the beam might be somewhat larger than 1 cm, FWHM; then for a given beam power, the peak heat flux would decrease as r_o^{-2}. According to my analysis, $\Delta T_{tritide} \sim r_o^{-2}$, $\Delta T_{Cu} \sim r_o^{-1.5}$, $\Delta T_{conv} \sim r_o^{-1}$, and $\Delta T_{caloric}$ is unchanged. Thus if the beam diameter (FWHM) were increased a mere 10% to 1.1 cm, we would have ΔT=284°C, which is below the 300°C threshold at which thermal decomposition would limit lifetime. In fact, measurements by Schumacher suggest the beam FWHM may be nearer to 1.5 cm, which would reduce ΔT to only 174°C. This seems by far the most likely explanation.

Second, my analytical approximations and assumptions could have introduced such a 12% error.

The third possibility is that the maximum tolerable peak surface temperature is in fact somewhat larger than 300°C, due to the cladding effect of the native titanium oxide. It is known [16] that the native oxides of many refractory metals are excellent barriers to hydrogen permeation. This may be occuring in the RTNS targets. Logan [6] reports that an experiment was once performed in which the titanium film

was clad with palladium without breaking vacuum, and then tritided. This would replace the native titanium oxide film with one of palladium (which is essentially transparent to the tritium). That target exhibited normal initial neutron fluxes but a noticeably (but not enormously) shorter lifetime. Without further data, it is difficult to say how effective a native oxide barrier might be.

3 Suggested RTNS Improvements

3.1 Tritide Layer Thermal Resistance Reduction

An improvement which could be tried almost immediately would be to decrease the thickness of the $TiT_{1.5}$ layer to 2.5 μm from its present value of 10 μm. As discussed above, calculations indicate that the deuteron range is only 1.87 μm, with a very small (~10%) standard deviation. This change would not reduce the neutron flux, and would decrease the effective thermal resistance by a factor of 6 to give $\Delta T_{tritide}$=31°C in the $TiT_{1.5}$ layer.

It is tempting to reduce the tritide layer thickness even further, to 1.6 μm. This would mean that essentially all the deuterons would pass completely through the $TiT_{1.5}$ layer, still carrying about 50 keV of energy, and then stop in the underlying substrate. Logan [8] has reported that implanting deuterium in or near the copper results in rapid target degradation. If, however, a layer of palladium (k=0.76 W/cm-K) were interposed between the tritide and the copper the target could be designed so that virtually all the deuterium would be implanted in the palladium, which readily supports hydrogen diffusion. The deuterium would thus rapidly diffuse from a peaked, narrow implant profile into a uniform distribution through the Pd. Furthermore, motivated by Ref. [17], one could devise techniques to allow the deuterium to escape through microscopic channels in the TiT_x layer. This could result in a very long-lived target, perhaps much longer than the present ones, despite the reduced tritium content due to the thinner TiT_x layer. The thermal conductivity of the Pd would not be a problem as a layer thickness of a mere 0.5 μm would be more than adequate to capture 99.9% of the low-energy deuterons.

It turns out that 375 keV is **not** the optimal deuteron energy from a thermal efficiency viewpoint. Table 1 tabulates the deuteron stopping power $S=\delta E/\delta x$ and the D-T reaction cross section σ as a function of incident energy (laboratory frame). From these data, we can calculate the range R_p and the neutron yield Y(E) per incident deuteron:

$$Y(E) = N_T \int_0^E \sigma(E')/S(E')\, dE' ,$$

where N_T is the atomic density of tritium. Note that at 375 keV incident deuteron energy, we predict $Y(E)=3.3\times10^{-5}$ for $TiT_{1.5}$; yet initial specific yields in the very best targets have been observed to be as high as 4.2×10^{-5} [4], 27% larger than predicted. This is undoubtedly due to the fact that a fresh target is nearly saturated with tritium, i.e. it is very nearly TiT_2 rather than $TiT_{1.5}$. However tritium/titanium

E (keV)	σ(barns)	S (keV/μm)	Path Length	Projected Range R_p (μm)	Range Straggle (μm)	Neutron Yield (10^{-5})	Yield/keV (10^{-7} /keV)
20	0.054	109	0.29	0.11	0.18	0.004	0.02
30	0.246	130	0.37	0.16	0.21	0.013	0.04
40	0.665	146	0.45	0.21	0.24	0.038	0.09
50	1.33	158	0.51	0.27	0.24	0.088	0.18
60	2.20	169	0.57	0.31	0.26	0.170	0.28
70	3.14	178	0.63	0.36	0.27	0.287	0.41
80	3.96	185	0.68	0.40	0.28	0.438	0.55
90	4.61	190	0.74	0.45	0.29	0.613	0.68
100	4.95	195	0.79	0.49	0.30	0.803	0.80
150	3.98	205	1.04	0.72	0.32	1.66	1.11
200	2.50	203	1.28	0.96	0.32	2.27	1.14
250	1.67	194	1.53	1.19	0.34	2.67	1.07
300	1.20	183	1.80	1.45	0.35	2.96	0.99
350	0.974	172	2.08	1.72	0.36	3.20	0.91
375	0.886	167	2.23	1.87	0.36	3.30	0.88
400	0.798	162	2.38	2.02	0.36	3.40	0.85
450	0.666	153	2.70	2.34	0.36	3.58	0.80
500	0.572	145	3.03	2.67	0.36	3.74	0.75

Table 1: $T(d,n)^4He$ neutron production yields for $TiT_{1.5}$ targets.

ratios in excess of 1.5 are not nearly as thermally stable as the dissociation pressure curves in Figure 3 clearly show. After a short period of operation, the target would outgas down to the stable plateau of $TiT_{1.5}$, at which time the dissociation rate would be as calculated in Section 2. This hypothesis is supported by reports [8, 18] of an initial, relatively rapid decay of 30% in neutron target yields, followed by a much more gradual decay.

The energy efficiency of the neutron generation process is Y(E)/E (also tabulated in Table 1), and this peaks at 200 keV, where it is 30% greater than at 375 keV. Coincidentally, a 200 kV, 20 A power supply is available at LLNL. Use of a beam generator energized by this supply would result in an across-the-board improvement, because the same beam power yields the same temperature rise, but now produces 1.3 times as many neutrons. (Of course the beam current must be nearly doubled in order to compensate for the reduced beam voltage.) Even better, the range of a 200-keV deuteron in $TiT_{1.5}$ is only 0.96 μm. This

would allow the use of a $TiT_{1.5}$ layer only 0.75 μm thick, backed by 0.5 μm of Pd. As shown in Table 1, in the last 0.2 μm of flight, the deuteron energy is too low to produce a significant number of neutrons. The thermal resistance of this $TiT_{1.5}$/Pd composite is 0.24 cm^2-°C/kW To produce the same number of neutrons as is realized at present, an 8°C temperature rise would be incurred in the tritide/Pd layer. Although there have been reports that the surface of the TiT_x layer is depleted of tritium, Logan [8] has shown that such regions must be less than 0.1 μm thick because the measured RTNS neutron yield at 200 keV is in agreement with predictions

Yet another scheme to reduce the thermal resistance of the tritide layer would be to fabricate microscopic copper "fingers" poking through the tritide layer. These could be fabricated by photoetching, followed by chemical vapor deposition of the titanium layer to fill the grooves. Alternatively the titanium could be evaporated and the surface mechanically polished For high-aspect ratio grooves, the temperature rise would be reduced by approximately the ratio

$$T_{new}/T_{old}=(w_c+w_w)k_{TiT_x}/w_w k_{Cu}$$

where w_c is the width of the tritide region and w_w is the width of the copper fingers. However, the neutron flux will be reduced by the factor $w_c/(w_c+w_w)$. As such this technique is only attractive when the thermal resistance of the tritide layer is the dominant term in that case we can reduce it fivefold at the expense of a 25% reduction in neutron flux by optimal exploitation of this approach. Because of this, the copper fingers are a desirable option if the proposed order-of-magnitude thinning of the tritide layer proves impractical. If, however, we can apply the other procedures described above to reduce the tritide thickness to less than 1 μm, then its thermal resistance is negligible relative to the other terms and the use of copper fingers would be the wrong approach

3.2 Convective Thermal Resistance

The convective component of the peak RTNS target temperature can be lowered substantially by reducing the thermal time constant τ_{conv}. As discussed in Section 2.3, the present RTNS system has $\tau_{conv} \gg \tau_{rev}$, where τ_{rev} is the target period of rotation. This implies that the convective heat transfer may be correctly analyzed in terms of the average heat flux $<q^*>$ If τ_{conv} is made less than τ_{rev}, however, then most of the heat will drain out of the copper before the next thermal impulse arrives. Thus the copper will start from a near-ambient temperature, which results in a temperature profile of the form $\Delta T_{Cu}(z)=\Delta T^0_{Cu}\ e^{-z/z_0}$ in thick copper Consider now the case where the copper layer is so thin that its thickness H is comparable to z_0; now we can no longer use the preceding simple formula. Assume the other side of the copper is a relative thermal insulator (a conservative and reasonable approximation for the cooling channels). The situation is equivalent to having a thick copper layer with an infinite array of image heat sources at $z=\pm 2nH$. Thus $\Delta T_{Cu}(z)=\Delta T^0_{Cu}(e^{-z/z_0}+e^{(z-2H)/z_0})/(1-e^{-2H/z_0})$. The peak temperature rise at the surface of the copper is thus

$$\Delta T_{Cu} = \Delta T^{o}_{Cu} \cdot \coth H/z_o \qquad (1)$$

Now the temperature will quickly equilibrate (in time $H^2/\alpha \ll \tau_{rev}$, where α is the thermal diffusivity) to a uniform value throughout this copper layer; this value is $<T(t=0)> = (\Delta T^{o}_{Cu})(1-e^{-2H/z_o})(z_o/H)$. The temperature will now decay with a time constant $\tau_{conv}=RC$, where $C=\rho C_{Cu} H$ and $R=(1/h)(A_{target}/A_{channel})$ where h is the convective heat transfer coefficient for the channel flow and $A_{channel}/A_{target}$ is the ratio of heat-transfer (channel) surface area to target surface area. So after a single revolution, we have

$$<T(t=\tau_{rev})> = <T(t=0)> \cdot e^{-\tau_{rev}/\tau_{conv}}.$$

This is the temperature of the copper layer just before the next beam thermal pulse is added. After several revolutions, the temperature will stabilize at

$$\Delta T_{conv} = <T(t=\infty)> = \exp(-\tau_{rev}/\tau_{conv}) + \exp(-2\tau_{rev}/\tau_{conv}) + \ldots =$$

$$= <T(t=0)>/(\exp(\tau_{rev}/\tau_{conv}) - 1) .$$

Thus

$$\Delta T_{conv} = \Delta T^{o}_{Cu} \cdot \frac{1-e^{-2H/z_o}}{(H/z_o)(e^{\tau_{rev}/\tau_{conv}} - 1)}, \text{ where } \Delta T^{o}_{Cu} = 0.17q(k\rho C r_o^{3} V)^{-1/2} . \qquad (2)$$

We are interested in finding the value of H which minimizes the sum of ΔT_{conv} and ΔT_{Cu}. For present target parameters, this works out to be H=210 μm, for which $(\Delta T_{conv}+\Delta T_{Cu})$ is 126°C. By redesigning the channel geometry and flow conditions, we could further reduce the convective thermal resistance. For example, one could increase the cooling water to 1200 psi from its present value of 300 psi. This does not greatly change the mechanical design of the target, because the target is already handling 1400 psi of hydrostatic pressure at the rim (due to centrifugal force). Increasing the supply pressure to 1200 psi would only increase the rim hydrostatic pressure by 450 psi, i.e. a 32% increase. The channel lengths could also be reduced by a factor of 2 because it is unlikely that the innermost portion of an RTNS target would be usable in an upgraded system due to its much lower velocity. The cooling channels could have the dimensions indicated in Figure 4, i.e. 300 μm wide, on 500 μm centers, and 600 μm deep. These changes would reduce both the convective and caloric thermal resistances by a factor of 3.

If the changes suggested in Sections 3.1 and 3.2 were performed, the following temperature rises would now result from generating the same neutron flux as described in Section 2 (recall that the total beam power has been reduced by a factor of 1.3 because of the increased neutronic efficiency at 200 keV):

TiT_x/Pd:	$\Delta T_{tritide} = 8°C$
Copper:	ΔT_{Cu} =76°C
Water (boundary layer):	ΔT_{conv} =7°C
Water (caloric heating):	$\Delta T_{caloric}$ =4°C
Total:	ΔT =95°C

Thus these relatively simple changes would allow us to increase the RTNS neutron flux by more than a factor of 3 over presently attainable levels. However, this assumes that ΔT is limited to only 300°C, which need not be the case for a "clad" target, as discussed in the following section.

3.3 Cladding of the Target Material

It has long been known [16] that metal hydrides can be clad with materials having very low hydrogen permeability, enabling them to exist at far higher temperatures than would otherwise be possible. As an example, clad zirconium hydride has been used in nuclear reactors at 1150°C, at which temperature unprotected $ZrH_{1.5}$ would be completely dissociated (dissociation pressure of ~90 atmospheres). This would be equivalent to operating the TiT_x at 890°C, which is far higher than is presently possible. This 890°C is also a maximum reasonable temperature in view of its proximity to the melting point of copper (1083°C). This would allow generation of neutron fluxes 9 times that of present targets, when combined with the previous suggestions. Of course the cladding layers would have to be quite thin ($\leq$0.1 μm) so as not to slow the incident deuterons too much.

There are a number of materials which appear to be virtually impermeable to high-pressure tritium at 890°C, even at thicknesses of only 0.1 μm. For example, permeation data compiled by Steve Steward [19] indicate that a 0.1 μm-thick silicon layer would confine the tritium under these conditions for over 10,000 hours, provided the layer is free from pinholes. Tungsten would also work, and quite possibly so would TiN or TiO (suggested in Ref. [17]).

The TiT_x will need to be clad on all surfaces. Furthermore it is desirable to partition the TiT_x into a large number of small, individually clad islands. This is to reduce the target's vulnerability to pinholes in the cladding; a single pinhole would affect only the neutron production from its own island, not its neighbors. The narrow spaces between the islands could be used as outlets for the deuterium which would be implanted into the underlying Pd layer. Figure 4 is a sketch of how this might look. This structure could easily be fabricated using a sequence of conventional microlithography and thin-film deposition and etching techniques, such as are used in integrated circuit fabrication.

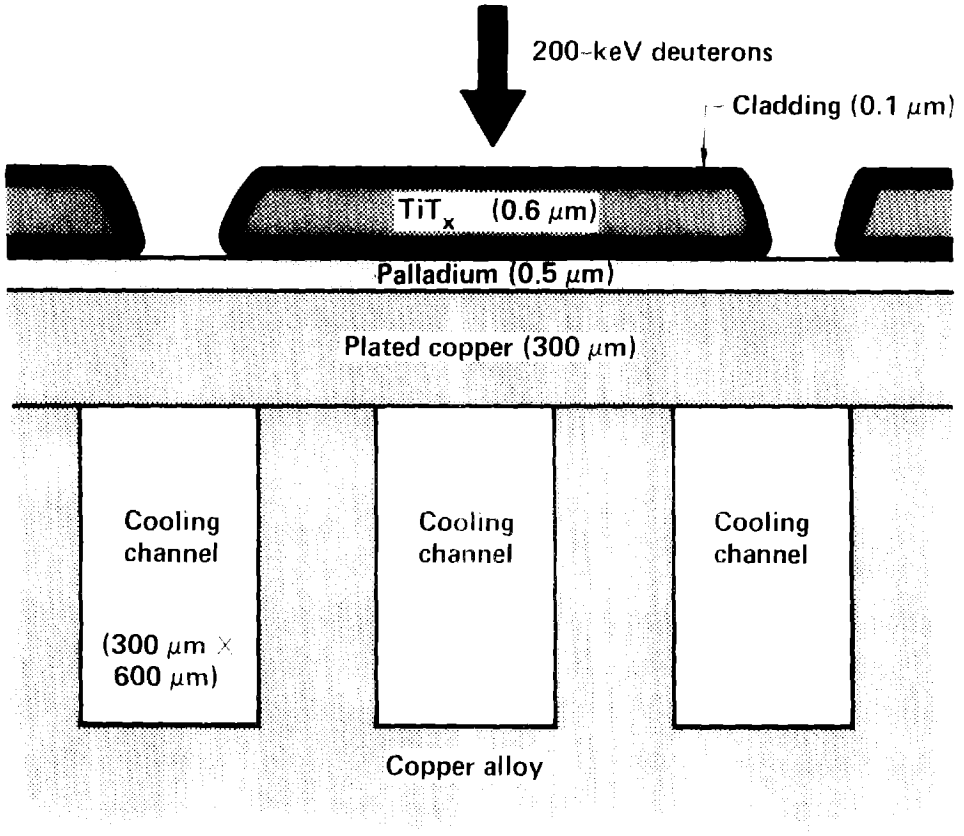

Figure 4: Cross section of the proposed upgraded RTNS target.

3.4 Increased Target Velocity

After implementing the above target modifications, the thermal resistance is virtually all due to the thermal resistance of the copper thermal diffusion zone underlying the TiT_x/Pd layers. This thermal resistance is proportional to $V^{-1/2}$, so a doubling of target velocity V would permit a 40% increase beam current density, hence in neutron flux. This could be achieved either by doubling the rotation rate or by doubling the target diameter. This would not be an easy modification in view of the rotating vacuum seal and water supply, but it should be possible, as airplane propellors routinely operate at stress levels such as would be generated by such a target upgrade (e.g. 2850 RPM for a 2-meter diameter). Increasing the diameter of the central flange (the water supply fitting) by a factor of 2 would simplify the mechanical design without encroaching too much on the tritided target area. Logan [6] has pointed out that a flat target (rather than a spherical cap) would be much better suited to handle the mechanical stresses induced by an increased target velocity. Of course a control system would be required to maintain the desired sample clearance as the target chamber is tilted.

3.5 Integral Heat Pipes

In lieu of increasing target velocity, one could fabricate integral heat pipes within the copper target substrate in order to transport more heat in the vertical (depth) direction than can be obtained solely by thermal diffusion into the copper. For example, if one fabricated in the copper deep (200 μm) narrow ($\simeq$30 μm) nickel-plated ducts, partially filled with a metallurgically compatible liquid (perhaps mercury), then the metal would vaporize and transport heat at sonic speeds down the channel. Condensation could occur during the remainder of the target's rotation period. The ducts could be angled slightly to enable the centrifugal potential to force the liquid back to its original location at the top of the duct. This process is very difficult to analyze and would have to be numerically modeled. Howeever it seems likely that such a design could accomodate at least a 40% increase in heat flux. As such it may be a more economical approach than doubling the target velocity.

3.6 Reduced target clearance

As mentioned earlier, the peak neutron flux right at the TiT_x layer is 60% greater than at the minimum sample distance of 0.35 cm. At present, this fixed clearance is preset when the target is stationary. The clearance cannot presently be reduced, due to the thickness of the target (1.5 mm), target runout, and target flattening during rotation. However, the sample could be moved much closer to the target if a control system were installed to track the target as it is scanned radially. I estimate that at least a 25% increase in peak neutron flux could be achieved with such a system; the full 60% cannot be achieved as it would require a perfectly formed, infinitesimally thin target.

3.7 Corrugated Targets

In order to produce yet higher neutron fluxes, one could fashion corrugations in the the target in the direction of the incident beam [20]. This is sketched in Figure 5 for a 3:1 aspect ratio of the corrugations. This reduces the peak heat flux seen by the copper, which is the primary thermal limit in an optmized design. It is important that the copper layer be at least 215 μm thick, because that is the depth to which the majority of the heat is confined when the beam passes over it (see Section 3.2). It is also important that there not be large flat regions between the peaks and valleys, because these regions would absorb the full beam flux at normal incidence and hence would overheat. The cooling water flows inside the corrugations.

The aspect ratio of this scheme is limited by the maximum temperature of the peaks of the corrugations, because the heat transfer is poor there. The only constraint is that the copper not be destroyed; it does not greatly matter if the TiT_x is absent at the tops of the corrugations. (It actually is permissible for the copper peaks to melt momentarily, as the melted region will be confined to a depth of $\sim$100 μm.) If the target is not clad, then the TiT_x temperature is limited to 300°C and there is a factor of 5 in benefit to be

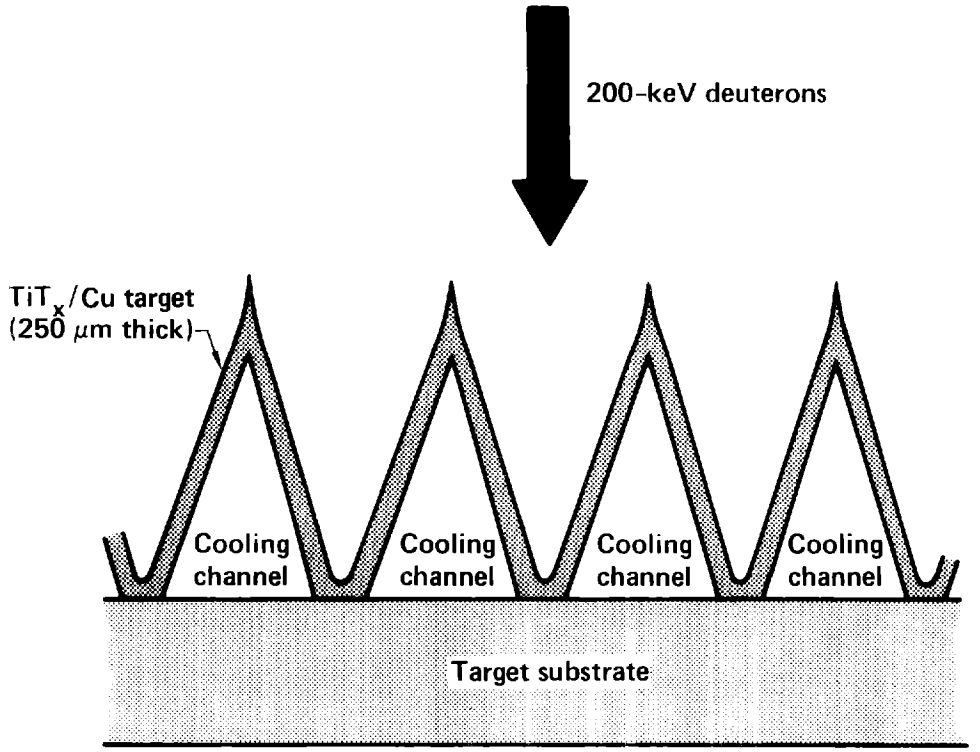

Figure 5: Proposed corrugated target surface.

had by corrugating the copper with a 5:1 aspect ratio. However, if the cladding procedure is successful, then the TiT_x can already be operated near the melting point of the copper, so it seems that only a factor of 2 beam flux increase (with a corresponding corrugation aspect ratio of 2) can be straightforwardly achieved without vaporizing the target. It is possible that higher aspect ratios may be achieved by clever design of the corrugation peaks; a logarithmic shape is desired. This would require further analysis and numerical modeling of the heat transfer and deuteron scattering near the corrugation peaks [20].

3.8 Summary of Upgraded Target Thermal Performance

The preceding set of RTNS target and system upgrade ideas have been sketched in Figs. 4 and 5. The multiplicative factors by which neutron peak flux can be increased as a consequence of each improvement are calculated below. I have listed them in recommended order of implementation, i.e. easiest ones first.

Modification	Peak neutron flux	Required beam current increase
Thinner TiT_x, Cu, and channel width:	×2.8	×2.8
Reduced target/sample clearance (control system):	×1.25	-----
Optimized beam voltage (200 kV):	×1.3	×1.9
Cladding of TiT_x:	×2.5	×2.5
Doubled target velocity **or** integral heat pipes	×1.4	×1.4
Corrugations (2:1 for clad targets):	×2.0 (est.)	×2.0
Net gain	×22	

It is likely that not all techniques will work out precisely as predicted, and that the implementations may interfere with each other to some extent. Nonetheless, a 10-fold increase in peak neutron flux should be readily achievable, with the possibility of much more if all the technological problems can be worked out (particularly for target corrugation). To realize a 10-fold gain in peak neutron flux, a 1.5 Ampere, 200 keV deuteron beam would be required. The ion source and transport system for such a beam is not a trivial problem, but is believed to be technically feasible [21]

3.9 Target Lifetime

It is desirable that the upgraded RTNS targets have a useful life comparable to or longer than the present lifetime of 100 hours, because neutron activation of the target room precludes rapid changing of the targets. Considering that the neutron output from present RTNS targets is only reduced by 30% of its initial value after receiving a deuterium dose 5 times that of the target's tritium content, it is somewhat surprising that the targets last as long as they do. This can perhaps be explained [22] by the fact that the deuterons are being implanted in a narrow, heavily damaged region and hence will be diffuse as interstitials through the TiT_x and back into the vacuum system without disturbing the tritium. This would work if the activation energy for breaking the Ti-T bond is greater than for interstitial diffusion of deuterons through the TiT_x (as seems likely).

The proposed upgraded targets would receive 10 or more times the neutron dose that the present targets receive, yet the total tritium content would be only 1/15 as much (because of the much thinner TiT_x layer). Thus it seems essential that the upgraded target be designed so that virtually all the deuterium passes through the TiT_x layer without stopping [17]. With an underlaying layer of Pd, the deuterium could then diffuse out of the target through slots in the TiT_x (Fig. 4) Damage to the TiT_x layer would be minimal as most ion implantation damage is generated near the end of an ion's range, where nuclear stopping processes dominate [23]. Damage to the Pd itself would of course occur, but this damage could be annealed out by using the D_2 beam to heat the target thus acting as a thermal pulse annealer. Physical sputtering of the target would not be a serious problem, because deuteron sputtering yields onto

Ti or Si targets are less than 2×10^{-3} ejected atoms/incident deuteron at 200 keV [24]. However, there may be implantation damage to the cladding due to Rutherford scattering, which could increase its permeability to tritium (radiation-enhanced diffusion). The probability of this could be minimized by orienting the cladding layers so as to promote "channeling". This needs further investigation.

If the inclusion of a Pd underlayer does not by itself yield the desired 100-hour lifetime, one could design an **in situ** retritiding system (no manual intervention required). Of course this technique would only work for targets in which the TiT_x has not been clad with a tritium-impermeable material. The rotation rate of the target would be slowed down so that the temperature in the TiT_x momentarily rises to ~700°C once each rotation, enough to drive out any hydrogen (implanted deuterium as well as bound tritium). The strength of the copper alloy target substrate would not be harmed because the heat pulse does not penetrate deeply enough. Any radiation damage to the titanium and palladium might be annealed by the thermal pulse. Then the D_2 beam would be shut off and the inside of the target chamber would be backfilled with tritium gas. By heating the target to 350°C (e.g. using a lamp or a blanket heater), the tritium would bond to the titanium just as in the initial target fabrication. The target is then ready for reuse.

4 Conclusions and Recommendations

4.1 Conclusions

This report has described techniques and designs by which the peak neutron flux from RTNS targets could in principle be boosted by a factor of up to 30. To be conservative, an upgrade factor of 10 is recommended. A 1.5-Ampere, 200-kV ion source, accelerator, and beam transport system would be needed to realize such a 10-fold gain in peak neutron flux. The main ideas are:

1. The present RTNS targets would be maximally thermally efficient (i.e. produce 30% more neutrons per unit of beam power) at 200 keV beam energy, compared with the present 375 keV. By a happy coincidence, a 200 kV, 20 A power supply exists at LLNL and is available for use.

2. The heat flux in present targets is primarily limited by the thermal resistance of the TiT_x layer (presently 10 μm thick). This term can be greatly reduced by thinning the TiT_x layer to 2.5 μm for the present 375 keV beam energy; at the recommended 200 keV energy, the TiT_x need only be 1 μm thick. An array of copper "fingers", roughly a micron wide, could be fabricated in the target surface to reduce the thermal resistance of the TiT_x if it is impractical to thin the TiT_x layer to 1 μm thickness.

3. Convective heat transfer presently involves significant thermal resistance. We can get a lower peak target temperature by thinning the outermost layer of copper from its present value of 500 μm to 215 μm (no thinner, lest the thermal capacitance of the copper layer begin to suffer). Further improvements can be achieved by spacing the cooling channels much closer together, increasing the cooling channel aspect ratio, increasing the water supply pressure, and decreasing the cooling channel length.

4. If approaches 2 and 3 are implemented, the limiting thermal resistance becomes that of the copper. Improvements could be made by increasing the rotational target velocity V; the gains are proportional to $V^{1/2}$. Alternatively, integral heat could be fabricated within the copper target. Yet another approach would be to corrugate the target surface; this can be done profitably up to about a 5:1 aspect ratio for unclad targets, or 2:1 for clad targets, affording a commensurate increase in neutron flux. Higher aspect ratios may be feasible but will require a precise design of the corrugation peak shape.

5. Presently, the sample must be positioned at least 0.35 cm from the TiT_x layer. Ths could be reduced by implementing a control system to maintain minimum clearance, thus increasing the neutron flux. This may be necessary anyway if a flat (rather than spherical) target is used in the upgraded design.

6. Target lifetime will be a serious consideration but can probably be dealt with when we understand the degradation mechanisms better. It seems advantageous to design a palladium underlayer in which the deuterons stop, and then diffuse to exit regions which have been etched in the overlying TiT_x layer. It is also suggested that unclad targets may be retritided **in situ**, possibly by using the heating effect of the deuteron beam to evaporate trapped deuterium and as a thermal pulse annealer; then one could leak in a fresh supply of tritium gas. Alternatively, the use of materials to clad the TiT_x (as suggested below) may make this unnecessary

7. The present target is limited to a peak temperature of ~300°C. For $TiT_{1.5}$ targets, this temperature might be increased to as high as 890°C by using a suitable cladding layer such as silicon, tungsten, TiO_2 or TiN. This could also enhance target lifetime. The TiT_x should be partitioned into islands prior to cladding in order to protect the target against severely degraded whole-target performance due to a small number of pinholes in the cladding.

4.2 Recommendations

The following research and development program is a suggested sequence to achieve the target upgrade:

- In support of the proposed thinned TiT_x layer
    - Precise measurements of deuteron stopping distance in TiT_x, with particular attention paid to minimizing the number of deuterons which stop in the tritide layer (for enhanced lifetime). The possibility of channeling should also be considered; elimination of channeling may require knowledge of the TiT_x crystallite orientations.
    - Experimental measurement of the thin-film thermal conductivity of TiT_x as a function of temperature, in the form used on RTNS targets
    - Development of deposition techniques to allow uniform, accurate deposition of Ti and Pd (e.g. $0.8\mu m \pm 10\%$) over the entire target.
    - Development of lithographic and etching processes to fabricate titanium islands with a palladium underlayer.
- Development of a manufacturing process to make narrower copper channels of higher aspect ratio, capped by a thinner layer of plated copper then is presently used.

- Redesign of cooling water rotating seal for higher water flow.

- Lifetime studies:
  - Direct measurements of D_2 diffusion through TiT_x as a function of temperature.
  - Measurement of implant damage to TiT_x as a function of depth.
  - Measurement of damage to an underlying Pd layer in which deuterons stop; annealing experiments (if needed).
  - Measurement of target lifetime with an underlying Pd layer.
  - Development of on-line beam diameter and target temperature diagnostics (e.g. using infrared microscopy or by melting spots of a test material).

- Re-tritiding experiments:
  - Use of the deuteron beam to thermally drive out D_2 and possibly to anneal Ti, without weakening the underlying Cu alloy substrate.
  - Attempt to retritide an RTNS target (with and without the above preheat).
  - Development of gate valve and blanket heater to allow **in-situ** retritiding.

- Cladding studies:
  - Study of cladding materials compatibility (Si, W, TiO, TiN) with TiT_x.
  - Test of maximum temperature capability of clad TiT_x.
  - Measure effect of sustained D_2 ion implantation and sputtering damage on cladding layers.
  - Development of deposition techniques for achieving thin, uniform, pinhole-free cladding materials, preferable oriented so as to promote channeling.
  - Study pinholes in the cladding: their occurence and consequences (to determine how many TiT_x islands are required).
  - Develop lithographic processes for fabricating clad TiT_x islands.

- Target corrugation studies:
  - Detailed thermal modeling of corrugations; optimization of corrugation peak shape.
  - Development of fabrication technology for corrugated targets (especially a bonding technique).

- Design of target with doubled velocity:
  - Stress analyses of flat (rather than spherical) targets.
  - Development of control system to allow precise control of sample clearance from the target.

- Development of bonding techniques which can support channel pressures of ~5000 psi.
- Redesign of rotating vacuum and water seals, probably with larger flanges.

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